UNIVERSITI PUTRA MALAYSIA

EFFECT OF MAGNETIC ELEMENTS SUBSTITUTION ON THE INTERGRANULAR PROPERTIES OF BI-BASED SUPERCONDUCTORS

MALIK IDRIES ADAM

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MALIK IDRIES ADAM

Thesis submitted to the Senate of Universiti Putra Malaysia in fulfillment of the requirements for the degree of Doctor of Philosophy

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Abstract of thesis submitted to the Senate of Universiti Putra Malaysia in fulfillment of the requirements for the degree of Doctor of Philosophy

EFFECT OF MAGNETIC ELEMENTS SUBSTITUTION ON THE INTERGRANULAR PROPERTIES OF Bi-BASED SUPERCONDUCTORS

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May 2003

Chairman: **Professor Dr. Abdul Halim Shaari**

Faculty: **Science and Environmental Studies**

The effects of magnetic elements substitution for calcium position in the $Bi_{1.6}Pb_{0.4}Sr_2(Ca_{1-x}M_x)_2Cu_3O_\delta$ with M=Pr, Nd, Sm, Gd, Dy or Ni and x=0.0-0.10 system, prepared by the conventional solid state reaction method have been investigated by means of structural, transport and magnetic methods. The samples were sintered in air at 855°C for 150 hours followed by annealing at 830°C for 30 hours. The superconducting properties such as volume fraction of present phases, lattice parameters, grain shape and critical temperatures were affected by the introduction of different elements for Ca in the system. The volume fraction of the high-T_c(2223) phase decreases and that of the low-T_c(2212) phase increases with the increase of substituents. As a result, the c-axis parameter decreased from $37\,\mathring{A}$ to $30.7\,\mathring{A}$ at initial substitution stages for all samples. Thus, the critical temperature, T_c decreased to that of the low-T_c(2212) phase. The morphological aspects of the samples generally show platelet-like grains with

random orientations, typically for 2212 and 2223 phase's structure. Air annealing at 830°C for 30 hours slightly improved the physical properties of the samples.

Measurements of AC magnetic susceptibility as a function of temperature and ac field amplitudes have been carried out for all samples at constant frequency f=125Hz. The effect of magnetic elements substitution was then investigated in terms of AC susceptibility. It was found that as the amount of the substituent content increases, the diamagnetic onset temperature decreases for all samples. Temperature-dependent critical current density was deduced from the AC susceptibility data for some selected samples in order to compare their result. The analysis for comparison is based on the availability of higher percentage of the high-T_c(2223) phase in the sample, the suppression degree of diamagnetic behaviour with respect to ac fields; rapid or slow shift of the summit in $\chi''(T)$ to lower temperature with increasing field amplitude, and the sharpness of $\chi'(T)$ for intergranular component for the same field amplitude. Although intergranular critical current density was found to be high for Ni-and Nd-substituted samples, the intergranular critical current density generally decreased with the increase of the substituent content in (BiPb)-2223 system.

Frequency and ac field amplitude dependence in the range of 20-1000Hz of the low field AC susceptibility were also investigated for some selected samples. It is observed that the ac loss peaks slightly shift to higher

temperatures with increasing frequency. This behaviour can be interpreted as hysteretic bulk pinning losses as well as intergranular flux creep losses. The experimental results were qualitatively discussed in the framework of the critical state model. Consequently, the effective volume fraction of the grains and the temperature dependence of the intergranular critical current density were estimated by means of the best fit of the calculated data for the experimental matrix susceptibility. Since the granularity effect was very pronounced at low field range, the contribution of the grains and matrix were separated from the total measured AC susceptibility. The effective volume fraction of the grains was found to vary between 0.33 to 0.24 depending on the substituted element oxide.

The average activation energy as a function of frequency at various ac field amplitudes is found to be inversely proportional to the power of magnetic field. Among the samples studied, Ni-substituted sample (x =0.02) was found to increase the flux pinning of Bi(Pb)-2223 system. This was derived from the fact that the suppression degree of the diamagnetic behaviour is smallest for higher fields compared to other samples. On the other hand, the intergranular current dependent activation energy was deduced by fitting the intergranular peak temperature T_p versus field amplitude at frequency f=20Hz for all samples. The intergranular critical current density dependence of activation energy is found to obey $E_a \propto j^{-\mu}$ relation, which is a characteristic of the vortex glass/collective creep models, hence indicating that the intergranular weak links has a collective pinning behaviour inside the samples. The best pinning behaviour was found in Ni-substituted sample with x=0.02.

Abstrak tesis yang dikemukakan kepada Senat Universiti Putra Malaysia sebagai memenuhi keperluan untuk ijazah Doktor Falsafah

Kesan gantian unsur-unsur magnet ke atas ciri-ciri antara butiran untuk superkonduktor berasaskan Bismut

Oleh

MALIK IDRIES ADAM

Mei 2003

Pengerusi: **Profesor Dr. Abdul Halim Shaari**

Faculti: **Sains dan Pengajian Alam Sekitar**

Kesan gantian unsur-unsur bermagnet keatas kedudukan kalium di dalam sistem $Bi_{1.6}Pb_{0.4}Sr_2(Ca_{1-x}M_x)_2Cu_3O_\delta$ dengan M=Pr, Nd, Sm, Gd, Dy atau Ni dan x=0.0-0.1, yang disediakan menggunakan kaedah tindak balas biasa bagi keadaan pepejal telah dikaji melalui kaedah struktur, pengangkutan dan magnet. Sampel disinter di dalam udara pada suhu 855°C selama 150 jam diikuti dengan sepuhlindapan pada suhu 830°C selama 30 jam. Ciri-ciri kesuperkonduksian seperti peratus isipadu fasa-fasa yang wujud, parameter kekisi, bentuk butiran dan suhu genting semuanya dipengaruhi dengan mengganti unsur-unsur berbeza bagi Ca di dalam sistem tersebut. Peratus isipadu untuk fasa T_c-tinggi (2223) berkurangan dan peratus isipadu fasa T_c-rendah (2212) bertambah dengan penambahan peratus bahan gantian. Ini menatijahkan pengurangan parameter paksi-*c* dari $37\,\text{Å}$ ke $30.7\,\text{Å}$ pada peringkat awal penggantian bagi semua sampel. Oleh itu suhu genting T_c berkurangan ke nilainya untuk fasa T_c-rendah (2212). Aspek morfologi bagi sample-sampel ini biasanya menunjukkan butiran yang

berkepingan dengan orientasi rawak; tipical untuk struktur fasa 2212 dan 2223. Sepuh lindapan di dalam undara pada suhu 830°C selama 30 jam dapat meningkatkan sedikit sifat fizik sampel.

Ukuran bagi kerentanan magnet arus AU sebagai satu fungsi suhu dan magnitud medan AU telah dibuat untuk semua sampel pada frekuensi f=125Hz. Kesan gantian unsur-unsur magnet dikaji di dalam sebutan kerentanan magnet arus AU. Didapati bahawa apabila peratus unsur gantian bertambah, maka suhu permulaan kesan diamagnet berkurangan untuk semua sampel. Ketumpatan arus genting yang bergantung pada suhu dianggar dari data-data kerentanan magnet AU bagi beberapa sampel yang dipilih untuk membuat perbandingan. Analisis untuk perbandingan adalah berdasarkan peratus kewujudan fasa Tc-tinggi (2223) dalam sampel darjah pengurangan ciri diamagnet merujuk kepada medan AU, kelajuan peralihan puncak $\chi''(T)$ ke suhu rendah bila amplitud medan bertambah dan ketajaman $\chi'(T)$ untuk komponen antara butiran bagi medan dengan amplitud yang sama. Walaupun ketumpatan arus genting untuk sampel yang unsur gantiannya adalah Ni dan Nd, tetapi secara keseluruhan, ketumpatan arus genting antara butiran berkurangan dengan penambahan gantian bagi sistem (BiPb)-2223.

Kebergantungan kerentanan magnet AU untuk medan rendah keatas frekuensi dan amplitud medan AU telah dikaji di dalam julat frequensi 20-1000Hz untuk beberapa sampel. Didapati bahawa puncak kelesapan AU berpindah ke suhu lebih tinggi bila frekuensi bertambah. sifat ini boleh

difahamkan sebagai kesan kelesapan terpin pukal histerisis dan juga kelesapan disebabkan oleh pergerakan fluks antara butiran. Hasil eksperimen ini dibincangkan secara kualitatif di dalam rangka model keadaan genting. Anggaran peratusan isipadu berkesan untuk butiran dan penertuan kebergantungan kepada suhu bagi ketumpatan arus genting antara butiran diperolehi dengan cara *'fitting'* terbaik antara data-data eksperimen dan matrik kerentanan. Disebabkan kesan butiran amat ketara pada julat medan rendah, maka sumbangan butiran dan matrik diasingkan dari kerentanan arus AU yang diukur. Peratus isipadu berkesan didapati di antara 0.33 ke 0.24 bergantung kepada oksida unsur gantian.

Purata tenaga teraktif sebagai fungsi frekuensi bagi berbagai amplitud medan AU didapati berkadar songsang dengan kuasa medan magnet. Diantara sampel yang dikaji, sampel gantian Ni (x=0.002) didapati menambahkan "keterpinan fluks bagi sistem Bi (Pb)-2223. Ini diperolehi dari fakta bahawa darjah pengurangan ciri diamagnet adalah terkecil untuk medan lebih tinggi berbanding dengan sampel-sampel lain. Sebaliknya, kebergantungan arus antara butiran keatas tenaga keaktifan dianggar dari fitting puncak suhu antara butiran T_p melawan amplitud medan pada frekuensi f=20Hz untuk semua sampel. Kebergantungan ketumpatan arus genting antara butiran didapati mengikut pertalian $E_a \propto j^{-\mu}$, yang merupakan ciri untuk model kaca/rayapan kolektif yang menunjukkan hubungan lemah antara butiran mempunyai ciri terpin kolektif di dalam sampel. Ciri terpin terbaik didapati di dalam sampel gantian Ni dengan x=0.02.

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I certify that an Examination Committee met on May,27th,2003 to conduct the final examination of Malik Idries Adam on his Doctor of Philosophy thesis entitled "The Effect of Magnetic Elements Substitution on the Intergranular Properties of Bi-Based Superconductors" in accordance with Universiti Pertanian Malaysia (Higher Degree) Act 1980 and Universiti Pertanian Malaysia (Higher Degree) Act 1981. The committee recommends that the candidate be awarded the relevant degree. Members of the Examination Committee are as follows:

ELIAS SAION, Ph.D.
Associate Professor,
Faculty of Science and Environmental Studies
Universiti Putra Malaysia
(Chairman)

ABDUL HALIM SHAARI, Ph.D.
Professor,
Faculty of Science and Environmental Studies
Universiti Putra Malaysia
(Member)

KAIDA KHALID, Ph.D.
Professor,
Faculty of Science and Environmental Studies
Universiti Putra Malaysia
(Member)

ZAINUL ABIDIN HASSAN, Ph.D.
Faculty of Science and Environmental Studies
Universiti Putra Malaysia
(Member)

DON McKENZIE PAUL, Ph.D.
Professor,
Department of Physics, University of Warwick,
Coventry CV4 7AL, UK.
(Independent Examiner)

GULAM RUSUL RAHMAT ALI, Ph.D.
Professor/Deputy Dean
School of Graduate Studies
Universiti Putra Malaysia

Date: 21 JUL 2003

The thesis submitted to the Senate of Universiti Putra Malaysia has been accepted as fulfilment of the requirement for the degree of Doctor of Philosophy. The members of the Supervisory Committee are as follows:

ABDUL HALIM SHAARI, Ph.D.
Professor,
Faculty of Science and Environmental Studies
Universiti Putra Malaysia
(Chairman)

KAIDA KHALID, Ph.D.
Professor,
Faculty of Science and Environmental Studies
Universiti Putra Malaysia
(Member)

ZAINUL ABIDIN HASSAN, Ph.D.
Faculty of Science and Environmental Studies
Universiti Putra Malaysia
(Member)

AINI IDERIS, Ph.D.
Professor/Dean
School of Graduate Studies
Universiti Putra Malaysia

Date: 15 AUG 2003

DECLARATION

I hereby declare that the thesis is based on my original work except for quotations and citations, which have been duly acknowledged. I also declare that it has not been previously or concurrently submitted for any other degree at UPM or other institutions.

MALIK IDRIES ADAM

Date: 09th February 2003

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